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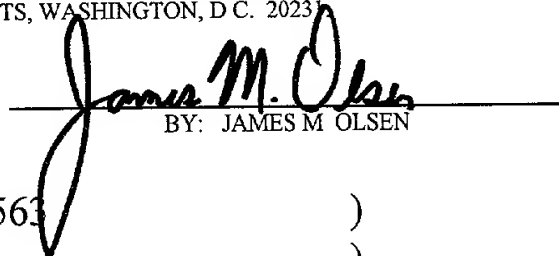
417 REC'D PCT/PTO 01 DEC 2000

PATENT  
Attorney Docket No. H 3491 PCT/US

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

CERTIFICATE OF EXPRESS MAIL  
(37 C.F.R. § 1.10)

I HEREBY CERTIFY THAT THIS PAPER OR FEE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE, EXPRESS MAIL POST OFFICE TO ADDRESSEE" UNDER 37 C.F.R. § 1.10, BEARING EXPRESS MAIL LABEL NO. **EK219526757US** ON THIS 1ST DAY OF DECEMBER, 2000 AND IS ADDRESSED TO: COMMISSIONER FOR PATENTS, WASHINGTON, D.C. 20231

  
BY: JAMES M. OLSEN

Int'l Appl. No.: PCT/EP99/03563 )  
Int'l Filing Date: May 25, 1999 )  
Priority Date: June 3, 1998 )  
Applicants: Beatrix KOTTWITZ et al. ) Art Unit: Unassigned  
Serial No.: To be assigned ) Examiner: Unassigned  
Filed: December 1, 2000 )  
For: DETERGENT CONTAINING AMYLASE )  
AND PERCARBONATE )

**Commissioner for Patents**  
**Box PCT**  
**Washington, D.C. 20231**

**TRANSMITTAL OF APPLICATION PAPERS TO U.S.  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. § 371 (37 CFR 1.494 OR 1.495)**

This transmittal letter is based upon Form PTO-1390.

The above-identified applicant has filed an International Application under the PCT and hereby submit(s) to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. [X] This is the FIRST submission of items concerning a filing under 35 U.S.C. §371.

## International Application No. PCT/EP99/03563

2. ☐ This is the SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. §371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay.
4. ☒ A proper Demand for International Preliminary Examination (IPE) was made to the appropriate Authority (IPEA) by the 19th month from the earliest claimed priority date (see attached).
5. ☒ A copy of the International Application as amended (35 U.S.C. §371(c)(2)) -  
(a) ☐ is transmitted herewith (required when not transmitted by International Bureau).  
(b) ☒ has been transmitted by the International Bureau.  
(c) ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into the English language is enclosed.
7. ☒ Amendments to the (specification and) claims of the International Application under PCT Article 19 (35 U.S.C. §371 (c)(3))  
(a) ☐ are transmitted herewith (required if not transmitted by the International Bureau).  
(b) ☐ have been transmitted by the International Bureau.  
(c) ☐ have not been made; however, the time limit for making such amendments has NOT expired.  
(d) ☒ have not been made and will not be made.  
(e) ☐ will be submitted with the appropriate surcharge.
8. ☐ A translation of the amendments to the claims (and/or the specification) under PCT Article 19 (35 U.S.C. §173(c)(3)) is enclosed or will be submitted with the appropriate surcharge.
9. ☒ An unexecuted Oath or Declaration/Power of Attorney of the inventor(s) (35 U.S.C. §371(c)(4)) is enclosed.
10. ☐ A translation of at least the Annexes to the IPE Report under PCT Article 36 (35 U.S.C. §371(c)(5)) is enclosed.

**International Application No. PCT/EP99/03563**

**Items 11. to 16. below concern other document(s) or information included:**

- 11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98 is enclosed.
- 12. ☐ An Assignment is enclosed for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31. is included.
- 13. ☒ A FIRST preliminary amendment is enclosed. **IT IS REQUESTED THAT THE FILING FEES FOR THE CLAIMS BE CALCULATED AFTER THE CLAIM AMENDMENTS IN THE PRELIMINARY AMENDMENT HAVE BEEN ENTERED.**
- 14. ☐ A substitute specification (including claims, abstract, drawing) is enclosed.
- 15. ☐ A change of Power of Attorney and/or address letter is enclosed.
- 16. ☒ Other items of information:
  - ☐ This application is being filed pursuant to 37 CFR 1.494(c) or 1.495(c), and any **missing parts** will be filed before expiration of -
  - ☐ 22 months from the priority date under 37 CFR 1.494(c), or
  - ☐ 32 months from the priority date under 37 CFR 1.495(c).
  - ☐ The undersigned attorney is authorized by the International application and by the inventors to enter the **National Phase** pursuant to 37 CFR 1.494(c) or 1.495(c).

The following additional information relates to the International Application:

- ☒ Receiving Office: EPO (originally filed in the German Patent Office)
- ☐ IPEA (if filing under 37 CFR 1.495): EPO
- ☒ Priority claim(s) (35 USC §§ 119, 365): App. No. 198 24 688.9  
Filed: June 3, 1998
- ☒ A copy of the International Search Report is
  - ☒ enclosed.
  - ☒ attached to the copy of the International Application.
- ☒ A copy of the Receiving Office Request Form is enclosed.

International Application No. PCT/EP99/03563

The fee calculation is set forth below.

# FEE CALCULATION

- [x] A check in payment of the filing fee, calculated as follows, is attached (37 CFR 1.492)

Basic Fee ..... \$860.00

Total Number of claims  
in excess of (20) times \$18 ..... \$0.00

Number of independent claims  
in excess of (3) times \$80 ..... \$0.00

Fee for multiple dependent  
claims \$270 ..... \$0.00

TOTAL FILING FEE ..... \$860.00

Kindly send us the official filing receipt.

The Commissioner is hereby authorized to charge any additional fees which may be required or to credit any overpayment to Deposit Account No. 03-2775. This is a "general authorization" under 37 CFR 1.25(b), except that no automatic debit of the issue upon allowance is authorized.

Respectfully submitted,

CONNOLLY BOVE LODGE & HUTZ LLP

Date: December 1, 2000

By: James M. Olsen  
James M. Olsen  
Reg. No. 40,408

PATENT  
Attorney Docket No. H 3491 PCT/US

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

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For: DETERGENTS CONTAINING )  
AMYLAASE AND PERCARBONATE )

**Commissioner for Patents**  
**Box PCT**  
**Washington, D.C. 20231**

Sir:

**PRELIMINARY AMENDMENT**

Prior to the examination of the above application, please amend this application as follows:

**IN THE CLAIMS:**

Please cancel claims 2-11, without prejudice or disclaimer of the subject matter thereof.

**IN THE ABSTRACT:**

Please add the attached Abstract of the Disclosure.

**REMARKS**

If there are any other fees due in connection with the filing of this response, please charge

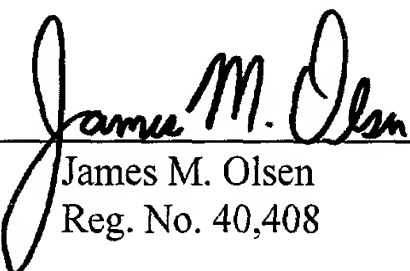
the fees to our Deposit Account No. 03-2775. If a fee is required for an extension of time under 37 C.F.R. § 1.136 not accounted for above, such an extension is requested and the fee should also be charged to our Deposit Account.

Respectfully submitted,

CONNOLLY BOVE LODGE & HUTZ LLP

Dated: December 1, 2000

By:

  
James M. Olsen  
Reg. No. 40,408

CONFIDENTIAL

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Attorney Docket No. H 3491 PCT/US (155\*314)

WO 99/63036

Detergents comprising amylase and percarbonate

The present invention relates to enzyme containing  
detergents comprising besides customary constituents an  
5 amylase from *Bacillus amyloliquefaciens* and a certain  
peroxidic bleach.

Laundry detergents, in addition to the ingredients  
vital to the laundering process, such as surfactants  
10 and builder materials, generally comprise further  
constituents, which may be grouped together under the  
heading of wash auxiliaries and which embrace such  
different active substance groups as foam regulators,  
graying inhibitors, bleaches, bleach activators, and  
15 color transfer inhibitors. Such auxiliaries also  
include substances which assist the surfactant  
performance by means of enzymatic degradation of stains  
present on the textile. The same applies, mutatis  
mutandis, to cleaning products for hard surfaces. As  
20 well as the proteases, which assist in removing  
proteins, and the fat-cleaving lipases, particular  
importance in this context attaches to the amylases.  
Amylases have the function of facilitating the removal  
of starchy stains by means of catalytic hydrolysis of  
25 the starch polysaccharide, and have been used for this  
purpose for a fairly long time in dishwashing  
detergents, but also in detergents for use in textile  
laundering. In by far the great majority of cases the  
amylase involved has comprised a heat-stable amylase  
30 from *Bacillus licheniformis*, which is customary in  
commerce under the designation Termamyl®, for example.  
More recently, there has been increased use in such  
compositions of genetically manipulated amylases; that  
is, amylases whose amino acid sequence has been  
35 altered, using gene technology methods, in comparison  
to naturally occurring amylases. As well as increasing  
their capacity to perform, the objective of genetically  
modifying amylases is essentially to enhance the  
stability of the enzyme, especially their stability to



attack by oxidizing agents. One approach toward achieving this objective, which was proposed in International Patent Application WO 94/18314, consists in removing particularly oxidation-susceptible amino acids, such as methionine, tryptophan, cysteine or tyrosine, from the amino acid sequence of the amylase, or replacing them by other amino acids more stable to oxidation. A similar approach is proposed in International Patent Application WO 95/21247, which recommends replacing at least one methionine in the amylase amino acid sequence by an amino acid which is neither methionine nor cysteine.

Although such genetic modifications may lead to improved amylase stability under certain application conditions, they do not help to increase the contribution of the amylase to the wash or cleaning performance of corresponding compositions comprising the amylase.

It has surprisingly now been found that the combination of a naturally occurring  $\alpha$ -amylase with a certain peroxidic oxidizing agent leads to unexpectedly synergistic performance improvements when used in detergents.

The invention accordingly provides an amylase containing detergent which comprises  $\alpha$ -amylase from *Bacillus amyloliquefaciens* and an alkali metal percarbonate, as well as customary ingredients compatible with such constituents.

The invention further provides for the use of such a combination for enhancing the cleaning performance of detergents, especially with respect to starchy and/or colored stains when used in detergent solutions, especially aqueous detergent solutions. In this context, cleaning performance with respect to colored stains is to be understood in its widest context and

embraces not only the bleaching of soil present on the textile and the bleaching of soil which is in the wash liquor, having become detached from the textile, but also the oxidative destruction of textile colors present in the wash liquor, having become detached from textiles under the wash conditions, before they are able to attach to different-colored textiles. Also, in the context of use in cleaning solutions for hard surfaces, this term refers both to the bleaching of soil present on the hard surface, especially tea, and to the bleaching of soil which is present in the dishwashing liquor, having become detached from the hard surface.

15  $\alpha$ -Amylase from *Bacillus amyloliquefaciens* has been known for a long time, for example, from the U.S. Patent US 1 227 374. It is available commercially, for example, under the designation Amylase BAN®.

20 A composition of the invention contains preferably from 0.001 mg to 0.5 mg, in particular from 0.02 mg to 0.3 mg, of amylolytic protein per gram of the overall composition. The protein concentration may be determined using known methods, such as the bicinchonic acid technique (BCA technique, Pierce Chemical Co., Rockford, IL) or the Biuret technique (A.G. Gornall, C.S. Bardawill and M.M. David, J. Biol. Chem. 177, 751-766, 1948).

30 A composition of the invention comprises preferably up to 50% by weight, in particular from 5% by weight to 30% by weight, of alkali metal percarbonate, sodium percarbonate being particularly preferred. It can be prepared by known processes and, especially for use in particulate compositions, may if desired be compounded in granular form and/or stabilized and/or coated, as known, for example, from International Patent Applications WO 91/15423, WO 92/17400, WO 92/17404, WO 93/04159, WO 93/04982, WO 93/20007, WO 94/03553,

WO 94/05594, WO 94/14701, WO 94/14702, WO 94/24044,  
WO 95/02555, WO 95/02672, WO 95/06615, WO 95/15291 or  
WO 95/15292 or in European Patent Applications  
EP 0 459 625, EP 0 487 256, EP 0 567 140, EP 0 623 553,  
5 EP 0 592 969 or EP 0 748 764. For reasons of stability  
it is preferably used in the form of granules which  
have been prepared from and/or coated with the aid of  
alkaline earth metal sulfate, alkali metal sulfate,  
alkali metal silicate, alkaline earth metal halide,  
10 alkali metal halide, alkali metal carbonate, alkali  
metal hydrogencarbonate, alkali metal phosphate, alkali  
metal borate, alkali metal perborate, boric acid,  
partially hydrated aluminosilicate, carboxylic acids,  
dicarboxylic acids, polymers of unsaturated carboxylic  
15 and/or dicarboxylic acids, or mixtures of these. In one  
preferred embodiment it has a morphology index (MI), as  
defined in EP 0 451 893, of below 0.06.

In one preferred embodiment of compositions of the  
20 invention, the ratio of  $\alpha$ -amylase to alkali metal  
percarbonate is in the range from 0.0001 mg to 0.1 mg,  
in particular from 0.001 mg to 0.01 mg, of amylolytic  
protein per % by weight of alkali metal percarbonate in  
the detergent.

25 In addition to the active substance combination used in  
accordance with the invention, the detergents of the  
invention, which may be present as solids, especially  
pulverulent solids, in postcompacted particle form, as  
30 homogeneous solutions or suspensions may in principle  
comprise all known ingredients which are customary in  
such compositions. The compositions of the invention  
may in particular comprise builder substances,  
surfactants, additional bleaches based on organic  
35 and/or inorganic peroxygen compounds, bleach  
activators, water miscible organic solvents, additional  
enzymes, sequesterants, electrolytes, pH regulators,  
and further auxiliaries, such as optical brighteners,  
graying inhibitors, color transfer inhibitors, foam

regulators, silver corrosion inhibitors, and also dyes and fragrances.

5 The compositions of the invention may comprise a surfactant or two or more surfactants, suitable surfactants comprising in particular anionic surfactants, nonionic surfactants and mixtures thereof, but also cationic, zwitterionic and amphoteric surfactants.

10

Suitable nonionic surfactants are, in particular, alkyl glycosides and ethoxylation and/or propoxylation products of alkyl glycosides or linear or branched alcohols having in each case 12 to 18 carbon atoms in the alkyl moiety and from 3 to 12, preferably from 4 to 10, alkyl ether groups. It is also possible to use corresponding ethoxylation and/or propoxylation products of N-alkylamines, vicinal diols, fatty acid esters and fatty acid amides, which in terms of the alkyl moiety correspond to the aforementioned long chain alcohol derivatives, and also alkylphenols having 5 to 12 carbon atoms in the alkyl radical.

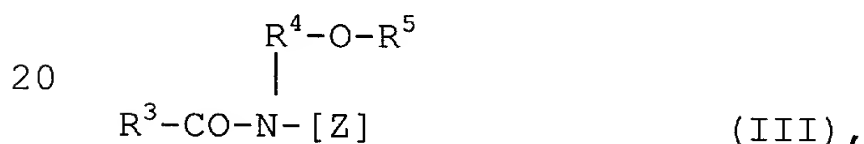
Nonionic surfactants used are preferably alkoxyated, advantageously ethoxylated, especially primary, alcohols having preferably 8 to 18 carbon atoms and on average from 1 to 12 mol of ethylene oxide (EO) per mole of alcohol, in which the alcohol radical may be linear or, preferably, methyl-branched in position 2 and/or may comprise linear and methyl-branched radicals in a mixture, as are commonly present in oxo alcohol radicals. In particular, however, preference is given to alcohol ethoxylates containing linear radicals from alcohols of natural origin having 12 to 18 carbon atoms, e.g., from coconut, palm, tallow fatty or oleyl alcohol and on average from 2 to 8 EO per mole of alcohol. Preferred ethoxylated alcohols include, for example, C<sub>12-14</sub> alcohols containing 3 EO or 4 EO, C<sub>9-11</sub> alcohols containing 7 EO, C<sub>13-15</sub> alcohols containing

3 EO, 5 EO, 7 EO or 8 EO, C<sub>12-18</sub> alcohols containing 3 EO, 5 EO or 7 EO, and mixtures thereof, such as mixtures of C<sub>12-14</sub> alcohol containing 3 EO and C<sub>12-18</sub> alcohol containing 7 EO. The stated degrees of ethoxylation represent statistical mean values, which for a specific product may be an integer or a fraction. Preferred alcohol ethoxylates have a narrowed homolog distribution (narrow range ethoxylates, NREs). In addition to these nonionic surfactants it is also possible to use fatty alcohols containing more than 12 EO. Examples thereof are (tallow) fatty alcohols containing 14 EO, 16 EO, 20 EO, 25 EO, 30 EO or 40 EO. Especially in detergents for use in machine dishwashing processes, it is common to use extremely low-foaming compounds. These include, preferably, C<sub>12-C18</sub> alkyl polyethylene glycol-polypropylene glycol ethers having in each case up to 8 mol of ethylene oxide and propylene oxide units in the molecule. However, it is also possible to use other nonionic surfactants which are known to be low-foaming, such as, for example, C<sub>12-C18</sub> alkyl polyethylene glycol-polybutylene glycol ethers having in each case up to 8 mol of ethylene oxide and butylene oxide units in the molecule, and also endgroup-capped alkyl polyalkylene glycol mixed ethers. Particular preference is also given to the hydroxyl-containing alkoxyated alcohols as are described in European Patent Application EP 0 300 305, referred to as hydroxy mixed ethers. The nonionic surfactants also include alkyl glycosides of the general formula RO(G)<sub>x</sub>, where R is a primary straight-chain or methyl-branched aliphatic radical, especially an aliphatic radical methyl-branched in position 2, containing 8 to 22, preferably 12 to 18, carbon atoms, and G represents a glucose unit having 5 or 6 carbon atoms, preferably glucose. The degree of oligomerization, x, which indicates the distribution of monoglycosides and oligoglycosides, is any desired number - which, as a variable to be determined analytically, may also be a fraction - between 1 and 10; preferably, x is from 1.2

to 1.4. Further suitable surfactants are polyhydroxy fatty acid amides of the formula (II)



where  $\text{R}^1\text{CO}$  is an aliphatic acyl radical having 6 to 22 carbon atoms,  $\text{R}^2$  is hydrogen or an alkyl or hydroxyalkyl radical having 1 to 4 carbon atoms, and [Z] is a linear or branched polyhydroxyalkyl radical having 3 to 10 carbon atoms and from 3 to 10 hydroxyl groups. The polyhydroxy fatty acid amides are derived preferably from reducing sugars having 5 or 6 carbon atoms, especially glucose. The group of the polyhydroxy fatty acid amides also includes compounds of the formula (III)



where  $\text{R}^3$  is a linear or branched alkyl or alkenyl radical having 7 to 12 carbon atoms,  $\text{R}^4$  is a linear, branched or cyclic alkylene radical or an arylene radical having 2 to 8 carbon atoms and  $\text{R}^5$  is a linear, branched or cyclic alkyl radical or an aryl radical or an oxyalkyl radical having 1 to 8 carbon atoms, preference being given to  $\text{C}_1\text{-C}_4$  alkyl radicals or phenyl radicals, and [Z] is a linear polyhydroxyalkyl radical whose alkyl chain is substituted by at least two hydroxyl groups, or alkoxyated, preferably ethoxyated or propoxyated, derivatives of said radical. Here too, [Z] is preferably obtained by reductive amination of a sugar such as glucose, fructose, maltose, lactose, galactose, mannose, or xylose. The N-alkoxy- or N-aryloxy-substituted compounds may then be converted to the desired polyhydroxy fatty acid amides, for example, in accordance with the teaching of International Patent Application WO 95/07331 by

reaction with fatty acid methyl esters in the presence of an alkoxide as catalyst. A further class of nonionic surfactants used with preference, which are used either as sole nonionic surfactant or in combination with other nonionic surfactants, in particular together with alkoxyated fatty alcohols and/or alkyl glycosides, are alkoxyated, preferably ethoxyated, or ethoxyated and propoxyated, fatty acid alkyl esters, preferably having 1 to 4 carbon atoms in the alkyl chain, especially fatty acid methyl esters, as are described, for example, in Japanese Patent Application JP 58/217598, or those prepared preferably by the process described in International Patent Application WO 90/13533. Nonionic surfactants of the amine oxide type, examples being N-cocoalkyl-N,N-dimethylamine oxide and N-tallowalkyl-N,N-dihydroxyethylamine oxide, and of the fatty acid alkanolamide type, may also be suitable. The amount of these nonionic surfactants is preferably not more than that of the ethoxyated fatty alcohols, in particular not more than half thereof. Further suitable surfactants include those known as gemini surfactants. This term is used generally to refer to those compounds which possess two hydrophilic groups per molecule. These groups are generally separated from one another as a result of what is known as a spacer. This spacer is generally a carbon chain, which should be long enough to give the hydrophilic groups a sufficient spacing to allow them to act independently of one another. Surfactants of this kind are generally notable for an unusually low critical micelle concentration and the ability to reduce greatly the surface tension of water. In exceptional cases, however, the expression gemini surfactants is used to embrace not only dimeric but also, correspondingly, trimeric surfactants. Examples of suitable gemini surfactants are sulfated hydroxy mixed ethers in accordance with German Patent Application DE 43 21 022 or dimer alcohol bis- and trimer alcohol tris-sulfates and ether sulfates in accordance with German Patent

Application DE 195 03 061. Endgroup-capped dimeric and trimeric mixed ethers in accordance with German Patent Application DE-A-195 13 391 are notable in particular for their bi- and multifunctionality. Thus said  
5 endgroup-capped surfactants possess good wetting properties and are low-foaming, so making them particularly suitable for use in machine washing or cleaning processes. However, it is also possible to use gemini-polyhydroxy fatty acid amides or polypolyhydroxy  
10 fatty acid amides, as described in International Patent Applications WO 95/19953, WO 95/19954, and WO 95/19955.

Suitable anionic surfactants are, in particular, soaps and those containing sulfate or sulfonate groups.  
15 Preferred surfactants of the sulfonate type are C<sub>9-13</sub> alkylbenzenesulfonates, olefinsulfonates, i.e., mixtures of alkenesulfonates and hydroxyalkanesulfonates, and also disulfonates, as are obtained, for example, from C<sub>12-18</sub> monoolefins having a terminal or  
20 internal double bond by sulfonating with gaseous sulfur trioxide followed by alkaline or acidic hydrolysis of the sulfonation products. Also suitable are alkanesulfonates, which are obtained from C<sub>12-18</sub> alkanes, for example, by sulfochlorination or sulfoxidation with  
25 subsequent hydrolysis or neutralization, respectively. Likewise suitable, in addition, are the esters of  $\alpha$ -sulfo fatty acids (ester sulfonates), e.g., the  $\alpha$ -sulfonated methyl esters of hydrogenated coconut, palm kernel or tallow fatty acids, which are prepared  
30 by  $\alpha$ -sulfonation of the methyl esters of fatty acids of plant and/or animal origin having 8 to 20 carbon atoms in the fatty acid molecule, followed by neutralization to give water-soluble mono-salts. Preferably, these comprise the  $\alpha$ -sulfonated esters of hydrogenated  
35 coconut, palm, palm kernel or tallow fatty acids, it being possible as well for sulfonation products of unsaturated fatty acids, e.g. oleic acid, to be present in small amounts, preferably in amounts of not more than about 2 to 3% by weight. Particular preference is



given to  $\alpha$ -sulfo fatty acid alkyl esters having an alkyl chain of not more than 4 carbon atoms in the ester group, examples being methyl esters, ethyl esters, propyl esters, and butyl esters. With particular advantage, the methyl esters of the  $\alpha$ -sulfo fatty acids (MES) are used, and also their saponified di-salts. Further suitable anionic surfactants are sulfated fatty acid glycerol esters which are the monoesters, diesters and triesters, and mixtures thereof, as obtained in the preparation by esterification of a monoglycerol with from 1 to 3 mol of fatty acid or in the transesterification of triglycerides with from 0.3 to 2 mol of glycerol. Preferred alk(en)yl sulfates are the alkali metal salts, and especially the sodium salts, of the sulfuric monoesters of  $C_{12}$ - $C_{18}$  fatty alcohols, examples being those of coconut fatty alcohol, tallow fatty alcohol, lauryl, myristyl, cetyl or stearyl alcohol, or of  $C_{10}$ - $C_{20}$  oxo alcohols, and those monoesters of secondary alcohols of this chain length. Preference is also given to alk(en)yl sulfates of said chain length which contain a synthetic straight-chain alkyl radical prepared on a petrochemical basis, these sulfates possessing degradation properties similar to those of the corresponding compounds based on fatty-chemical raw materials. From a detergents standpoint,  $C_{12}$ - $C_{16}$  alkyl sulfates and  $C_{12}$ - $C_{15}$  alkyl sulfates, and also  $C_{14}$ - $C_{15}$  alkyl sulfates, are particularly preferred. In addition, 2,3-alkyl sulfates, which may for example be prepared in accordance with US Patents 3,234,258 or 5,075,041 and obtained as commercial products from Shell Oil Company under the name DAN®, are suitable anionic surfactants. Also suitable are the sulfuric monoesters of the straight-chain or branched  $C_{7-21}$  alcohols ethoxylated with from 1 to 6 mol of ethylene oxide, such as 2-methyl-branched  $C_{9-11}$  alcohols containing on average 3.5 mol of ethylene oxide (EO) or  $C_{12-18}$  fatty alcohols containing from 1 to 4 EO. Preferred anionic surfactants further include the salts

of alkylsulfosuccinic acid, which are also referred to as sulfosuccinates or as sulfosuccinic esters and which constitute the monoesters and/or diesters of sulfosuccinic acid with alcohols, preferably fatty alcohols and especially ethoxylated fatty alcohols. Preferred sulfosuccinates comprise C<sub>8-18</sub> fatty alcohol radicals or mixtures thereof. Especially preferred sulfosuccinates contain a fatty alcohol radical derived from ethoxylated fatty alcohols which themselves represent nonionic surfactants. Particular preference is given in turn to sulfosuccinates whose fatty alcohol radicals are derived from ethoxylated fatty alcohols having a narrowed homolog distribution. Similarly, it is also possible to use alk(en)ylsuccinic acid containing preferably 8 to 18 carbon atoms in the alk(en)yl chain, or salts thereof. Further suitable anionic surfactants include fatty acid derivatives of amino acids, for example, of N-methyltaurine (taurides) and/or of N-methylglycine (sarcosides). Particular preference is given here to sarcosides and to the sarcosinates and, of these, especially the sarcosinates of higher fatty acids, which may be mono- or polyunsaturated, such as oleyl sarcosinate. Further suitable anionic surfactants are, in particular, soaps. Suitable soaps include in particular saturated fatty acid soaps, such as the salts of lauric acid, myristic acid, palmitic acid, stearic acid, hydrogenated erucic acid and behenic acid, and, in particular, mixtures of soaps derived from natural fatty acids, e.g., coconut, palm kernel, or tallow fatty acids. Together with these soaps, or as substitutes for soaps, it is also possible to use the known alkenylsuccinic salts.

The anionic surfactants, including the soaps, may be present in the form of their sodium, potassium or ammonium salts and also as soluble salts of organic bases, such as mono-, di- or triethanolamine. Preferably, the anionic surfactants are in the form of

their sodium or potassium salts, in particular in the form of the sodium salts.

5 In laundry detergents of the invention, surfactants are present in proportions of preferably from 5% by weight to 50% by weight, in particular from 8% by weight to 30% by weight, whereas compositions for cleaning hard surfaces, especially for the machine cleaning of kitchen- and tableware, have lower surfactant contents  
10 of up to 10% by weight, in particular up to 5% by weight, and preferably in the range from 0.5% by weight to 3% by weight.

15 A composition of the invention comprises preferably at least one water soluble and/or water insoluble, organic and/or inorganic builder. The water soluble organic builder substances include polycarboxylic acids, especially citric acid and sugar acids, monomeric and polymeric amino polycarboxylic acids, in particular  
20 methylglycinediacetic acid, nitrilotriacetic acid and ethylenediaminetetraacetic acid, and also polyaspartic acid, polyphosphonic acids, especially aminotris-(methylenephosphonic acid), ethylenediaminetetrakis-(methylenephosphonic acid) and 1-hydroxyethane-1,1-  
25 diphosphonic acid, polymeric hydroxy compounds such as dextrin, and also polymeric (poly)carboxylic acids, especially the polycarboxylates of European Patent EP 0 625 992 or of International Patent Application WO 92/18542 or of European Patent EP 0 232 202, which  
30 are obtainable by oxidizing polysaccharides or, respectively, dextrans; polymeric acrylic acids, methacrylic acids, maleic acids and copolymers thereof, which may also contain in copolymerized form small fractions of polymerizable substances without  
35 carboxylic acid functionality. The relative molecular mass of the homopolymers of unsaturated carboxylic acids is generally between 3000 and 200,000, that of the copolymers between 2000 and 200,000, preferably from 30,000 to 120,000, based in each case on free

acid. A particularly preferred acrylic acid-maleic acid copolymer has a relative molecular mass of from 30,000 to 100,000. Commercially customary products are, for example, Sokalan® CP 5, CP 10 and PA 30 from BASF.

5 Suitable though less preferred compounds of this class are copolymers of acrylic acid or methacrylic acid with vinyl ethers, such as vinyl methyl ethers, vinyl esters, ethylene, propylene and styrene, in which the acid fraction is at least 50% by weight. As water  
10 soluble organic builder substances it is also possible to use terpolymers containing as monomers two unsaturated acids and/or their salts and, as the third monomer, vinyl alcohol and/or an esterified vinyl alcohol or a carbohydrate. The first acidic monomer or  
15 salt thereof is derived from a monoethylenically unsaturated C<sub>3</sub>-C<sub>8</sub> carboxylic acid and, preferably, from a C<sub>3</sub>-C<sub>4</sub> monocarboxylic acid, especially from (meth)acrylic acid. The second acidic monomer or salt thereof may be a derivative of a C<sub>4</sub>-C<sub>8</sub> dicarboxylic  
20 acid, maleic acid being particularly preferred, and/or a derivative of an allylsulfonic acid substituted in position 2 by an alkyl or aryl radical. Such polymers may be prepared in particular by processes which are described in German Patent DE 42 21 381 and German  
25 Patent Application DE 43 00 772, and generally have a relative molecular mass of between 1000 and 200,000. Further preferred copolymers are those described in German Patent Applications DE 43 03 320 and DE 44 17 734, containing as monomers preferably  
30 acrolein and acrylic acid/acrylic acid salts, and/or vinyl acetate. The organic builder substances, especially for the preparation of liquid compositions, may be used in the form of aqueous solutions, preferably aqueous solutions with a strength of from 30  
35 to 50 percent by weight. All said acids are generally used in the form of their water soluble salts, especially their alkali metal salts.

Organic builder substances of this kind may be present, if desired, in amounts of up to 40% by weight, in particular up to 25% by weight, and preferably from 1% by weight to 8% by weight. Amounts close to the stated upper limit are used preferably in pastelike or liquid compositions of the invention, especially those containing water.

Suitable water soluble inorganic builder materials include, in particular, alkali metal silicates, alkali metal carbonates and alkali metal phosphates, which may be present in the form of their alkaline, neutral or acidic sodium or potassium salts. Examples thereof are trisodium phosphate, tetrasodium diphosphate, disodium dihydrogen diphosphate, pentasodium triphosphate, so-called sodium hexametaphosphate, oligomeric trisodium phosphate having degrees of oligomerization from 5 to 1000, especially from 5 to 50, and also the corresponding potassium salts and/or mixtures of sodium and potassium salts. Water insoluble, water dispersible inorganic builder materials used are, in particular, crystalline or amorphous alkali metal aluminosilicates, in amounts of up to 50% by weight, preferably not more than 40% by weight, and in liquid compositions in particular from 1% by weight to 5% by weight. Among these, preference is given to the crystalline sodium aluminosilicates of detergent quality, especially zeolite A, P and, if appropriate, X, alone or in mixtures, in the form for example of a cocrystallizate of zeolites A and X (Vegobond® AX, a commercial product from Condea Augusta S.p.A.). Amounts close to the stated upper limit are used preferably in solid, particulate compositions. Suitable aluminosilicates possess in particular no particles having a size of more than 30  $\mu\text{m}$ , and preferably consist at least 80% by weight of particles having a size below 10  $\mu\text{m}$ . Their calcium binding capacity, which may be determined in accordance with the information in German Patent DE 24 12 837, is

generally in the range from 100 to 200 mg of CaO per gram.

Suitable substitutes or partial substitutes for said aluminosilicate are crystalline alkali metal silicates, which may be present alone or in a mixture with amorphous silicates. The alkali metal silicates which can be used as builders in the compositions of the invention preferably have a molar ratio of alkali metal oxide to  $\text{SiO}_2$  of below 0.95, in particular from 1:1.1 to 1:12, and may be present in amorphous or crystalline form. Preferred alkali metal silicates are the sodium silicates, especially the amorphous sodium silicates, having a molar ratio  $\text{Na}_2\text{O}:\text{SiO}_2$  of from 1:2 to 1:2.8. Those with a molar ratio  $\text{Na}_2\text{O}:\text{SiO}_2$  of from 1:1.9 to 1:2.8 may be prepared by the process of European Patent Application EP 0 425 427. As crystalline silicates which may be present alone or in a mixture with amorphous silicates it is preferred to use crystalline phyllosilicates of the general formula  $\text{Na}_2\text{Si}_x\text{O}_{2x+1} \cdot y\text{H}_2\text{O}$ , where x, the so-called modulus, is a number from 1.9 to 22, in particular from 1.9 to 4, and y is a number from 0 to 33, and preferred values for x are 2, 3 or 4. Crystalline phyllosilicates which fall under this general formula are described, for example, in European Patent Application EP 0 164 514. Preferred crystalline phyllosilicates are those where x in the stated general formula adopts the values 2 or 3. In particular, both  $\beta$ - and  $\delta$ -sodium disilicates ( $\text{Na}_2\text{Si}_2\text{O}_5 \cdot y\text{H}_2\text{O}$ ) are preferred, with  $\beta$ -sodium disilicate, for example, being obtainable by the process described in International Patent Application WO 91/08171.  $\delta$ -Sodium silicates having a modulus of between 1.9 and 3.2 may be prepared in accordance with Japanese Patent Applications JP 04/238 809 and JP 04/260 610. In addition, virtually anhydrous crystalline alkali metal silicates of the abovementioned general formula wherein x is a number from 1.9 to 2.1, prepared from amorphous alkali metal silicates, and preparable as described in European

Patent Applications EP 0 548 599, EP 0 502 325 and EP 0 452 428, may be used in compositions of the invention. In a further preferred embodiment of compositions of the invention, a crystalline sodium phyllosilicate having a modulus of from 2 to 3 is used, as may be prepared from sand and soda by the process of European Patent Application EP 0 436 835. Crystalline sodium silicates having a modulus in the range from 1.9 to 3.5, as are obtainable by the processes of European Patents EP 0 164 552 and/or EP 0 294 753, are used in a further preferred embodiment of compositions of the invention. Crystalline sheetlike silicates of the abovementioned formula are sold by Clariant GmbH under the trade name Na-SKS, e.g., Na-SKS-1 ( $\text{Na}_2\text{Si}_{22}\text{O}_{45} \cdot x\text{H}_2\text{O}$ , kenyaite), Na-SKS-2 ( $\text{Na}_2\text{Si}_{14}\text{O}_{29} \cdot x\text{H}_2\text{O}$ , magadiite), Na-SKS-3 ( $\text{Na}_2\text{Si}_8\text{O}_{17} \cdot x\text{H}_2\text{O}$ ) or Na-SKS-4 ( $\text{Na}_2\text{Si}_4\text{O}_9 \cdot x\text{H}_2\text{O}$ , makatite). Of these, those particularly suitable include Na-SKS-5 ( $\alpha\text{-Na}_2\text{Si}_2\text{O}_5$ ), Na-SKS-7 ( $\beta\text{-Na}_2\text{Si}_2\text{O}_5$ , natrosilite), Na-SKS-9 ( $\text{NaHSi}_2\text{O}_5 \cdot 3\text{H}_2\text{O}$ ), Na-SKS-10 ( $\text{NaHSi}_2\text{O}_5 \cdot 3\text{H}_2\text{O}$ , kanemite), Na-SKS-11 ( $\text{t-Na}_2\text{Si}_2\text{O}_5$ ) and Na-SKS-13 ( $\text{NaHSi}_2\text{O}_5$ ), but especially Na-SKS-6 ( $\delta\text{-Na}_2\text{Si}_2\text{O}_5$ ). A review of crystalline phyllosilicates is given, for example, by the articles published in "Hoechst High Chem Magazin 14/1993" on pages 33-38 and in "Seifen-Öle-Fette-Wachse, Vol. 116, No. 20/1990" on pages 805-808. In one preferred embodiment of compositions of the invention, use is made of a granular compound of crystalline phyllosilicate and citrate, of crystalline phyllosilicate and abovementioned (co)polymeric polycarboxylic acid, as is described, for example, in German Patent Application DE 198 19 187, or of alkali metal silicate and alkali metal carbonate, as is described, for example, in International Patent Application WO 95/22592 or as is available commercially, for example, under the name Nabion® 15.

Builder substances may be present in the compositions of the invention, if desired, in amounts of up to 90% by weight. They are preferably present in amounts of up

to 75% by weight. Laundry detergents of the invention have builder contents of in particular from 5% by weight to 50% by weight. In compositions of the invention for cleaning hard surfaces, especially for the machine cleaning of kitchen- and tableware, the amount of builder substances is in particular from 5% by weight to 88% by weight, preferably no water insoluble builder materials being used in such compositions. In a preferred embodiment of compositions of the invention intended in particular for the machine washing of kitchen- and tableware, from 20% by weight to 40% by weight of water soluble organic builder, especially alkali metal citrate, from 5% by weight to 15% by weight of alkali metal carbonate, and from 20% by weight to 40% by weight of alkali metal disilicate are present.

Additional peroxygen compounds that may be suitable for use in compositions of the invention include, in particular, organic peracids or peracidic salts of organic acids, such as phthalimidopercaproic acid, perbenzoic acid or salts of diperdodecanedioic acid, hydrogen peroxide, and inorganic salts which give off hydrogen peroxide under the washing conditions, including perborate, percarbonate, persilicate and/or persulfate such as Caroate. Where solid peroxygen compounds are additionally to be used, they may be employed in the form of powders or granules, which may also have been coated in a manner which is known in principle. The addition of small amounts of known bleach stabilizers such as, for example, of phosphonates, borates and/or metaborates and metasilicates, and also magnesium salts such as magnesium sulfate, may also be judicious here.

Bleach activators which may be used are compounds which under perhydrolysis conditions give rise to aliphatic peroxo carboxylic acids having preferably 1 to 10 carbon atoms, in particular 2 to 4 carbon atoms, and/or



substituted or unsubstituted perbenzoic acid. Suitable substances are those which carry O-acyl and/or N-acyl groups of the stated number of carbon atoms, and/or substituted or unsubstituted benzoyl groups. Preference is given to polyacylated alkylenediamines, especially tetraacetylenediamine (TAED), acylated triazine derivatives, especially 1,5-diacetyl-2,4-dioxohexahydro-1,3,5-triazine (DADHT), acylated glycolurils, especially tetraacetylglycoluril (TAGU), N-acyl imides, especially N-nonanoylsuccinimide (NOSI), acylated phenolsulfonates, especially n-nonanoyl- or isononanoyloxybenzenesulfonate (n- or iso-NOBS), carboxylic anhydrides, especially phthalic anhydride, acylated polyhydric alcohols, especially triacetin, ethylene glycol diacetate, 2,5-diacetoxy-2,5-dihydrofuran, and the enol esters known from German Patent Applications DE 196 16 693 and DE 196 16 767, and also acetylated sorbitol and mannitol and/or the mixtures thereof (SORMAN) described in European Patent Application EP 0 525 239, acylated sugar derivatives, especially pentaacetylglucose (PAG), pentaacetylfructose, tetraacetylxylose and octaacetyllactose, and acetylated, optionally N-alkylated glucamine and gluconolactone, and/or N-acylated lactams, for example, N-benzoylcaprolactam, which are known from International Patent Applications WO 94/27970, WO 94/28102, WO 94/28103, WO 95/00626, WO 95/14759 and WO 95/17498. The hydrophilically substituted acylacetals known from German Patent Application DE 196 16 769 and acyllactams described in German Patent Application DE 196 16 770 and in International Patent Application WO 95/14075 are likewise used with preference. The combinations of conventional bleach activators known from German Patent Application DE 44 43 177, may also be used. Such bleach activators may, especially when the abovementioned hydrogen peroxide donating bleaches are present, be present in customary quantities, preferably in amounts of from 0.5% by weight to 10% by weight, and in particular from

1% by weight to 8% by weight, based on overall composition, but are preferably all absent when the percarboxylic acid essential to the invention is used as the sole bleach.

5

In addition to the conventional bleach activators or instead of them it is also possible for the sulfonylmines and/or bleach boosting transition metal salts or transition metal complexes, known from  
10 European Patents EP 0 446 982 and EP 0 453 003, to be present as so-called bleaching catalysts, preferably in an amount of up to 1% by weight, in particular from 0.001% by weight to 0.25% by weight.

15 Enzymes which may be used in the compositions in addition to the amylase that is essential to the invention include those from the class of the proteases, lipases, cutinases, pullulanases, hemicellulases, cellulases, oxidases, laccases and  
20 peroxidases, and mixtures thereof. If desired, amylases other than the amylase essential to the invention may be present in addition to it. Particularly suitable active enzymatic substances are those obtained from fungi or bacteria, such as *Bacillus subtilis*, *Bacillus*  
25 *licheniformis*, *Bacillus lentus*, *Streptomyces griseus*, *Humicola lanuginosa*, *Humicola insolens*, *Pseudomonas pseudoalcaligenes*, *Pseudomonas cepacia* or *Coprinus cinereus*. The amylase essential to the invention and the enzymes that may be used in addition may - as  
30 described, for example, in European Patent EP 0 564 476 or in International Patent Application WO 94/23005 - be adsorbed on carrier substances and/or embedded in coating substances in order to protect them against premature inactivation. In the detergents of the  
35 invention they are present preferably in amounts of up to 5% by weight, in particular from 0.2% by weight to 4% by weight. Where the composition of the invention comprises protease, it preferably has a proteolytic activity in the range from about 100 PU/g to about

10,000 PU/g, in particular from 300 PU/g to 8000 PU/g. Where two or more enzymes are to be used in the composition of the invention, this may be accomplished by incorporating the two or more separate, or  
5 conventionally separately compounded enzymes, or by means of two or more enzymes compounded together in granules, as known, for example, from International Patent Applications WO 96/00772 or WO 96/00773.

10 The organic solvents which may be used besides water in the compositions of the invention, especially if they are present in liquid or paste form, include alcohols having 1 to 4 carbon atoms, especially methanol, ethanol, isopropanol and tert-butanol, diols having 2  
15 to 4 carbon atoms, especially ethylene glycol and propylene glycol, and also mixtures thereof and the ethers derivable from the aforementioned classes of compound. Water miscible solvents of this kind are present in the compositions of the invention preferably  
20 in amounts of not more than 30% by weight, in particular from 6% by weight to 20% by weight.

Additionally, the compositions may comprise further constituents customary in detergents. These optional  
25 constituents include, in particular, enzyme stabilizers, graying inhibitors, color transfer inhibitors, foam inhibitors, and optical brighteners and also dyes and fragrances. In order to protect against silver corrosion, silver corrosion inhibitors  
30 may be used in dishwashing detergents of the invention. A cleaning product of the invention for hard surfaces may, furthermore, comprise abrasive constituents, especially from the group consisting of quartz flours, wood flours, polymer flours, chalks and glass  
35 microbeads, and mixtures thereof. Abrasives are present in the detergents of the invention preferably at not more than 20% by weight, in particular from 5% by weight to 15% by weight.

In order to adjust the pH to a desired level which does not come about of itself through the mixing of the other components, the compositions of the invention may comprise system-compatible and environmentally compatible acids, especially citric acid, acetic acid, tartaric acid, malic acid, lactic acid, glycolic acid, succinic acid, glutaric acid and/or adipic acid, or else mineral acids, especially sulfuric acid, or bases, especially ammonium hydroxides or alkali metal hydroxides. pH regulators of this kind are present in the compositions of the invention in amounts of preferably not more than 20% by weight, in particular from 1.2% by weight to 17% by weight.

The color transfer inhibitors suitable for use in laundry detergents of the invention include, in particular, polyvinylpyrrolidones, polyvinylimidazoles, polymeric N-oxides such as poly(vinylpyridine N-oxide), and copolymers of vinylpyrrolidone with vinylimidazole.

Graying inhibitors have the function of keeping the soil detached from the textile fiber in suspension in the liquor. Suitable for this purpose are water soluble colloids, usually organic in nature, examples being starch, glue, gelatin, salts of ether carboxylic acids or ether sulfonic acids of starch or of cellulose, or salts of acidic sulfuric esters of cellulose or of starch. Water soluble polyamides containing acidic groups are also suitable for this purpose. Furthermore, starch derivatives other than those mentioned above may be used, aldehyde starches, for example. Preference is given to cellulose ethers, such as carboxymethylcellulose (Na salt), methylcellulose, hydroxyalkylcellulose and mixed ethers, such as methylhydroxyethylcellulose, methylhydroxypropylcellulose, methylcarboxymethylcellulose, and mixtures thereof, in amounts, for example, of from 0.1 to 5% by weight, based on the compositions.

As optical brighteners, laundry detergents of the invention may comprise derivatives of diaminostilbene-disulfonic acid and/or its alkali metal salts. Suitable, for example, are salts of 4,4'-bis(2-anilino-4-morpholino-1,3,5-triazinyl-6-amino)stilbene-2,2'-disulfonic acid or compounds of similar structure which instead of the morpholino group carry a diethanolamino group, a methylamino group, an anilino group, or a 2-methoxyethylamino group. It is also possible for brighteners of the substituted diphenylstyryl type to be present, for example, the alkali metal salts of 4,4'-bis(2-sulfostyryl)biphenyl, 4,4'-bis(4-chloro-3-sulfostyryl)biphenyl, or 4-(4-chlorostyryl)-4'-(2-sulfostyryl)biphenyl. Mixtures of the aforementioned optical brighteners may also be used.

Especially for use in machine processes, it may be of advantage to add customary foam inhibitors to the compositions. Examples of suitable foam inhibitors are soaps of natural or synthetic origin having a high C<sub>18</sub>-C<sub>24</sub> fatty acid fraction. Examples of suitable nonsurfactant-type foam inhibitors are organopolysiloxanes and their mixtures with microfine, optionally silanized silica and also paraffins, waxes, microcrystalline waxes, and mixtures thereof with silanized silica or bis-fatty acid alkylene diamides. With advantages, use is also made of mixtures of different foam inhibitors, for example, mixtures comprising silicones, paraffins, or waxes. The foam inhibitors, especially those containing silicone and/or paraffin, are preferably bound on a granular, water soluble or dispersible support substance. Particular preference is given to mixtures of paraffins and bisstearylethylenediamide.

The production of solid compositions of the invention presents no difficulties and may be done conventionally, for example, by spray drying or granulation, with the enzymes and any other heat

sensitive ingredients such as bleaches, for example, being added separately later on if desired. To produce compositions of the invention of heightened bulk density, in particular in the range from 650 g/l to 950 g/l, preference is given to a process known from European Patent EP 0 486 592 which includes an extrusion step. Another preferred preparation, with the aid of a granulation process, is described in European Patent EP 0 642 576.

To produce compositions of the invention in tablet form, which may have one or more phases, may have one or more colors, and consist in particular of one layer or of two or more layers, in particular of two layers, it is preferred to follow a procedure in which all of the constituents - per one layer if appropriate - are mixed with one another in a mixer and the mixture is compressed by means of conventional tableting presses, for example, eccentric presses or rotary presses, at pressing forces in the range from about 50 to 100 kN, preferably from 60 to 70 kN. Especially in the case of multilayer tablets it may be of advantage if at least one layer is compressed beforehand. This is preferably accomplished at pressing forces of between 5 and 20 kN, in particular from 10 to 15 kN. In this way, tablets which are fracture resistant and yet dissolve sufficiently quickly under application conditions are obtained without problems, having fracture strengths and flexural strengths of normally from 100 to 200 N, but preferably above 150 N. A tablet produced in this way preferably has a weight of from 10 g to 50 g, in particular from 15 g to 40 g. The three-dimensional form of the tablets is arbitrary and may be circular, oval or angular, with intermediate forms also being possible. Corners and edges are advantageously rounded. Circular tablets preferably have a diameter of from 30 mm to 40 mm. In particular, the size of tablets of angular or cuboid design which are introduced predominantly by way of the dosing apparatus of, for

example, the dishwashing machine is dependent on the geometry and on the volume of this dosing apparatus. Embodiments which are preferred by way of example have a basal area of (from 20 to 30 mm) × (from 34 to 40 mm), in particular of 26×36 mm or of 24×38 mm.

Liquid or paste detergents of the invention in the form of solutions comprising customary solvents are generally prepared by simply mixing the ingredients, which may be introduced in bulk or in solution into an automatic mixer.

#### Examples

##### Example 1

To determine the washing power, cotton fabrics soiled with standardized test stains were washed at 40°C (detergent dosing 76 g; water hardness 16°d [German hardness]; load 3.5 kg, short program) in a domestic washing machine (Miele® W 701). Washing was carried out using a detergent **V1** containing 0.25% by weight of noninventive amylase granules (Termamyl® 60T) and 0.25% by weight of protease granules (activity 200,000 PE/g) containing the *Bacillus lentus* protease and 18% by weight of sodium percarbonate and 5% by weight of TAED, a detergent **V2** of otherwise the same composition but containing, instead of Termamyl®, the protein-equivalent amount of the genetically modified amylase Duramyl®, a detergent **V3** of otherwise the same composition as **V1** but containing, instead of Termamyl®, the protein-equivalent amount of the genetically modified amylase Purafect® OxAm, a detergent **V4** of otherwise the same composition as **V1** but containing, instead of the Termamyl®, the protein-equivalent amount of the fungal amylase Fungamyl®, and an inventive detergent **M1** of otherwise the same composition as **V1** but containing, instead of Termamyl®, the protein-

equivalent amount of the amylase from *Bacillus amyloliquefaciens*.

The composition of the invention had a wash performance which was markedly superior to that of the compositions containing a different amylase. The situation is essentially the same when the percarbonate in the composition of the invention was replaced by perborate.

10 Example 2

Detergents (**V5** and **V6**, respectively) for the machine washing of kitchen- and tableware, comprising 55% by weight sodium tripolyphosphate (calculated as anhydrous), 4% by weight amorphous sodium disilicate (calculated as anhydrous), 22% by weight sodium carbonate, 9% by weight sodium percarbonate, 2% by weight TAED, 2% by weight nonionic surfactant, and also 1% or 2% by weight amylase granules (Termamyl® 60T) and 1.4% by weight protease granules (activity 200,000 PU/g) comprising the *Bacillus lentus* protease (remainder to 100% by weight water, perfume and dye), compositions (**V7** and **V8**, respectively), whose composition was otherwise like that of **V5** and **V6**, respectively, but which instead of Termamyl® contained protein-equal amounts of Duramyl®, and also compositions in accordance with the invention (**M2** and **M3**, respectively), whose composition was otherwise like that of **V5** and **V6**, respectively, but which instead of Termamyl® contained protein-equal amounts of amylase from *Bacillus amyloliquefaciens*, were tested as indicated below:

In a Miele® G 575 dishwasher (additions of 20 g of each test composition in the universal program, water hardness 14-16°dH [German hardness], operating temperature 55°C), in each case 6 plates soiled with standardized starch stains or bleachable stains were washed and the remaining stain residue was determined



gravimetrically and related to the initial value prior to washing (i.e., 100%). The compositions of the invention were significantly superior to the noninventive compositions in cleaning performance.

- 5 Essentially the same is true if the percarbonate in the compositions of the invention was replaced by perborate.

Annex to our letter of 05.29.2000  
PCT/EP99/03563 (H 3491 PCT)

Replacement page

- 27 -

What is claimed is:

1. An amylase containing detergent, characterized in that it comprises  $\alpha$ -amylase from *Bacillus amyloliquefaciens* and alkali metal percarbonate in addition to customary ingredients compatible with such constituents, and does not comprise any antibody directed against the amylase.
2. The composition as claimed in claim 1, characterized in that it contains amylase in amounts of from 0.001 mg to 0.5 mg, in particular from 0.02 mg to 0.3 mg, per gram of the overall composition.
3. The composition as claimed in claim 1 or 2, characterized in that it contains up to 50% by weight, in particular from 5% by weight to 30% by weight, of alkali metal percarbonate.
4. The composition as claimed in any of claims 1 to 3, characterized in that it contains from 0.5% by weight to 10% by weight, in particular from 1% by weight to 8% by weight, of compound which under perhydrolysis conditions gives off peroxycarboxylic acid.
5. The composition as claimed in any of claims 1 to 4, characterized in that it contains bleach boosting transition metal salts or transition metal complexes in an amount of up to 1% by weight, in particular from 0.001% by weight to 0.25% by weight.

6. The composition as claimed in any of claims 1 to 5, characterized in that the alkali metal percarbonate comprises sodium percarbonate.

7. The composition as claimed in any of claims 1 to 6, characterized in that the alkali metal percarbonate is used in the form of granules prepared and/or coated with the aid of alkaline earth metal sulfate, alkali metal sulfate, alkali metal silicate, alkaline earth metal halide, alkali metal halide, alkali metal carbonate, alkali metal hydrogencarbonate, alkali metal phosphate, alkali metal borate, alkali metal perborate, boric acid, partially hydrated aluminosilicate, carboxylic acids, dicarboxylic acids, polymers of unsaturated carboxylic and/or dicarboxylic acids, or mixtures of these.

8. The composition as claimed in any of claims 1 to 7, characterized in that the alkali metal percarbonate has a morphology index of below 0.06.

9. The composition as claimed in any of claims 1 to 8, characterized in that it contains from 0.0001 mg to 0.1 mg, in particular from 0.001 mg to 0.01 mg, of  $\alpha$ -amylase per percent by weight of alkali metal percarbonate.

10. The use of a combination of  $\alpha$ -amylase from *Bacillus amyloliquefaciens* and alkali metal percarbonate for enhancing the cleaning performance of detergents which do not comprise any antibody directed against the amylase, when used in washing and cleaning solutions, especially aqueous washing and cleaning solutions.

Replacement page

- 29 -

11. The use as claimed in claim 10, characterized in that the cleaning performance is enhanced in relation to starchy and/or colored stains.

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Patent and Trademark Office**DECLARATION FOR  
UTILITY OR DESIGN  
PATENT APPLICATION**☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial FilingAttorney Docket  
Number

H 3491 PCT/US

First Named  
Inventor

KOTTWITZ, Beatrix

COMPLETE IF KNOWN

Application Number

Filing Date

Group Art Unit

Examiner Name

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**DETERGENT CONTAINING AMYLASE AND PERCARBONATE**

(Title of the Invention)

the specification of which

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) 05/25/1999 as United States Application Number or PCT InternationalApplication Number PCT/EP99/03563 and was amended on (MM/DD/YYYY)                      (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES NO
198 24 688.9	Germany	06/03/1998	<input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>	<input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/> <input checked="" type="checkbox"/>

☐ Additional foreign application numbers are listed on a supplemental priority sheet attached hereto:

I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.
  	  	<input type="checkbox"/>

Burden Hour Statement: This form is estimated to take .4 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.

## DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP99/03563	05/25/1999	

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

☐ Firm Name  Customer Number or label   
OR  
☒ List Attorney(s) and/or agent(s) name and registration number below:

Name	Registration Number	Name	Registration Number
Wayne C. Jaeschke Glenn E. J. Murphy Stephen D. Harper	21,062 33,539 33,243	See supplemental sheet attached	

☒ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☐ Customer Number or label  OR ☒ Fill in correspondence address below

Name	James M. Olsen				
Address	Connolly, Bove, Lodge & Hutz				
Address	P.O. Box 2207 (1220 Market Street)				
City	Wilmington	State	DE	ZIP	19899
Country	USA	Telephone	302-658-9141	Fax	302-656-9072

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this unsigned

Given Name	Beatrix	Middle Initial		Family Name	KOTTWITZ	Suffix e.g. Jr.	
Inventor's Signature	Beatrix Kottwitz				Date	Dec. 12, 2000	
Residence: City	Duesseldorf	State		Country	Germany	Citizenship	Germany
Post Office Address	Urdenbacher Allee 51						
Post Office Address							
City	40593 Duesseldorf	State		Zip		Country	Germany
Applicant Authority							

☒ Additional inventors are being named on supplemental sheet(s) attached hereto

## DECLARATION

ADDITIONAL INVENTOR(S)  
Supplemental Sheet

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventorGiven  
Name**Horst-Dieter**Middle  
InitialFamily  
Name**SPECKMANN**Suffix  
e.g. Jr.Inventor's  
Signature*Horst-Dieter Speckmann*

Date

*Dec. 12, 2000*Residence:  
City**Langenfeld**

State

Country

**Germany**

Citizenship

**Germany**

Post Office Address

**Eichenfeldstrasse 24 a**

Post Office Address

City

**40764 Langenfeld**

State

Zip

Country

**Germany**Applicant  
Authority

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventorGiven  
Name**Karl-Heinz**Middle  
InitialFamily  
Name**MAURER**Suffix  
e.g. Jr.Inventor's  
Signature*Karl-Heinz Maurer*

Date

*Dec. 12, 2000*Residence:  
City**Erkrath**

State

Country

**Germany**

Citizenship

**Germany**

Post Office Address

**Dechanstrasse 5**

Post Office Address

City

**40699 Erkrath**

State

Zip

Country

**Germany**Applicant  
Authority

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventorGiven  
Name**Christian**Middle  
InitialFamily  
Name**NITSCH**Suffix  
e.g. Jr.Inventor's  
Signature*Christian Nitsch*

Date

*Dec. 12, 2000*Residence:  
City**Duesseldorf**

State

Country

**Germany**

Citizenship

**Germany**

Post Office Address

**Otto-Hahn-Strasse 185**

Post Office Address

City

**40591 Duesseldorf**

State

Zip

Country

**Germany**Applicant  
Authority

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventorGiven  
NameMiddle  
InitialFamily  
NameSuffix  
e.g. Jr.Inventor's  
Signature

Date

Residence:  
City

State

Country

Citizenship

Post Office Address

Post Office Address

City

State

Zip

Country

Applicant  
Authority

Additional inventors are being named on supplemental sheet(s) attached hereto

Attorney Docket Number: H 3491  
First Named Inventor: Beatrix KOTTWITZ  
Title: DETERGENT CONTAINING AMYLASE AND  
PERCARBONATE

Supplemental Sheet of Additional Attorney(s) and/or Agent(s) :

Connolly, Bove, Lodge & Hutz, of Wilmington, DE 19899

Rudolf E. Hutz	Reg. No. <u>22,397</u>
John D. Fairchild	Reg. No. <u>19,756</u>
Harold Pezzner	Reg. No. <u>22,112</u>
Richard M. Beck	Reg. No. <u>22,580</u>
Paul E. Crawford	Reg. No. <u>24,397</u>
Patricia Smink Rogowski	Reg. No. <u>33,791</u>
Robert G. McMorrow, Jr.	Reg. No. <u>30,962</u>
Ashley I. Pezzner	Reg. No. <u>35,646</u>
William E. McShane	Reg. No. <u>32,707</u>
Mary W. Bourke	Reg. No. <u>30,982</u>
Gerard M. O'Rourke	Reg. No. <u>39,794</u>
James M. Olsen	Reg. No. <u>40,408</u>
Francis DiGiovanni	Reg. No. <u>37,310</u>
Eric J. Evain	Reg. No. <u>42,517</u>
Daniel C. Mulveny	Reg. No. <u>45,897</u>
Elliot C. Mendelson (Agent)	Reg. No. <u>42,878</u>
Christine M. Hansen	Reg. No. <u>40,634</u>

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